

H₂O diffusion in silicate glasses and melts

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Diffusion of H₂O in silicate melts and glasses plays a critical role in bubble growth that powers explosive volcanic eruptions and in stability of glass. Although extensive work has been carried out, there are still some major gaps in our understanding of the diffusion process. This work is aimed at understanding how the diffusivity of H₂O depends on the anhydrous composition, the pressure, and the dissolved total H₂O concentration (H₂O_t).

Samples used in this study include KS and Rhy (natural obsidian glass from Mono Craters, California, USA; initial H₂O_t 0.8% by weight for KS and 1.9% for Rhy), NSL (natural peralkaline rhyolitic glass from New Zealand, with 2.0% H₂O_t), Ab (synthetic albite glass with 1.8% initial H₂O_t), AOQ (synthetic haplogranitic glass with nominal composition of Qz28Ab38Or34 containing either 1.3% H₂O_t or 2.2%), T3 (synthetic glass with nominal composition of Qz50An50 containing 2.0% H₂O_t). Table 1 lists the composition of the samples measured by electron microprobe.

Dehydration experiments were carried out at 584–605°C and 0.5 to 5 kbar Ar pressure in a rapid-quench cold-seal furnace. After heating of 16 to 73 hours, each quenched experimental wafer is sectioned to obtain a slice from which the H₂O diffusion profile is measured by infrared spectroscopy. The Bruker IFS88 IR spectrometer with microscope (A-590) setup at Hannover is used with a slit width of 5 to 25 µm. The H₂O_t concentration is obtained by summing up the OH and molecular H₂O

(H₂O_m) species concentrations measured at 4520 cm⁻¹ and 5230 cm⁻¹ combination bands.

All H₂O_t concentration profiles are well fit by assuming that H₂O_t diffusivity ($D_{H_2O_t}$) is proportional to H₂O_t (Fig. 1). They can also be fit well by assuming that molecular H₂O is the diffusing species and $D_{H_2O_m}$ is independent of H₂O_t. These results are consistent with both Zhang *et al.* (1991) and Nowak and Behrens (1997) since H₂O_t is relatively low. The effect of different fitting procedures to the resulting $D_{H_2O_t}$ is examined and found to be within 20% relative.

Experimental data show that $D_{H_2O_t}$ decreases with increasing pressure. For AOQ composition at 600 °C and 1% H₂O_t, $D_{H_2O_t}$ decreases from 0.25 µm²/s at 0.5 kbar, to 0.133 µm²/s at 2 kbar, and to 0.080 µm²/s at 5 kbar. The activation volume for H₂O diffusion inferred from the data is 17 ± 8 (2σ error) cm³/mol, somewhat greater than but still in agreement within experimental error with that inferred from high temperature diffusivity data for AOQ (5 to 11 cm³/mol, Nowak and Behrens, 1997).

The dependence of $D_{H_2O_t}$ on anhydrous composition is examined at 5 kbar Ar pressure and 600°C. $D_{H_2O_t}$ at 1 wt.% H₂O_t is 0.152, 0.080, 0.070, and 0.053 µm²/s for NSL, AOQ, Ab and KS/Rhy samples. For the T3 sample, the diffusion profile is too short to be measured, implying a maximum $D_{H_2O_t}$ at 1% H₂O_t of ~0.002 µm²/s, much less than that in other glasses. For comparison, $D_{H_2O_t}$ in SiO₂ glass (Moulson and Roberts, 1961) at 600°C and 1 bar is 0.044 µm²/s by a

TABLE 1. Anhydrous oxide concentrations (wt.%) of samples used

Sample	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	Sum
KS, Rhy	76.59	0.08	12.67	1.00	—	0.03	0.52	3.98	4.88	
NSL	75.45	0.19	10.05	4.29	0.12	0.00	0.17	5.27	4.56	100.10
Albite	69.19	0.00	18.83	0.00	0.00	0.00	0.00	11.96	0.02	100.00 [†]
AOQ	76.14	0.00	13.53	0.00	0.00	0.00	0.00	4.65	5.68	100.00 [†]
T3	71.64	0.00	18.48	0.00	0.00	0.00	10.13	0.00	0.00	100.00 [†]

[†] Normalized to 100%.

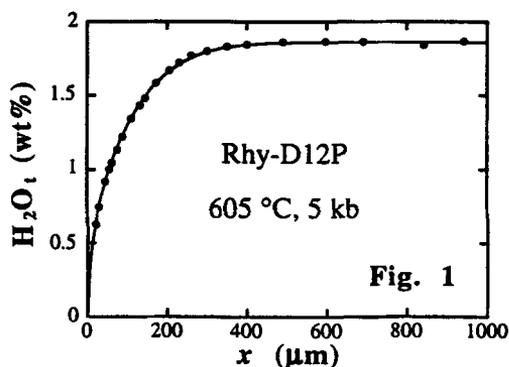


FIG. 1.

gross extrapolation to 1% H_2O_t , much greater than that in T3 glass even with the pressure effect being considered. Further, $D_{H_2O_t}$ in basaltic melts is significantly greater (Zhang and Stolper, 1991). We therefore conclude that (i) $D_{H_2O_t}$ values in fully polymerized and Ca-poor glasses and melts (AOQ, Ab and KS/Rhy) are not strongly dependent on the anhydrous composition, in agreement with Behrens and Nowak (1997); (ii) $D_{H_2O_t}$ increases with extra alkalis unassociated with Al (NSL) or with a decrease in degree of polymerization (basalt); (iii) assuming H_2O_m is the diffusing species, the greater $D_{H_2O_t}$ value in NSL is partially attributable to a greater H_2O_m concentration at a given H_2O_t compared with other samples, but also owing to a greater $D_{H_2O_t}$ and (iv) high Ca concentration in polymerized SiO_2 -rich melt reduces $D_{H_2O_t}$ (T3). The reduction of $D_{H_2O_t}$ by Ca may be due to complexing reactions between OH and Ca.

Both Ar solubility and diffusivity are obtained as a side product from this study since Ar is used as the pressure medium. The solubility of Ar in the glasses at 600°C and 5 kb is 0.52 wt.% in AOQ, 0.49% in KS/Rhy, 0.37% in NSL and 0.25% in Ab. The differences in the solubilities are small and consistent with other studies (e.g. White *et al.*, 1989). Examination of Ar diffusivity as a function of H_2O_t can reveal how the diffusivity of a molecular species depends on H_2O_t . This knowledge may be instructive to the understanding of H_2O_m diffusion. However, because Ar diffusion profile is relatively short (20 to 80 μm), loss of several μm glass from the edge during preparation and polishing of the sample has a significant effect on the measured profile. Hence, the effect of H_2O_t on Ar diffusion is not conclusive yet.

Work is in progress to investigate the effect of H_2O_t on $D_{H_2O_t}$ at higher H_2O_t content, and to study diffusion in dacitic and andesitic glasses and melts.

References

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